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1 **Metamorphic origin of anastomosing and wavy laminas overprinting putative microbial**

2 **deposits from the 3.22 Ga Moodies Group (Barberton Greenstone Belt)**

3

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21 **Keywords**

22 Paleoproterozoic; Barberton Greenstone Belt; Moodies Group; anastomosing and wavy laminae;
23 metasomatism

24

25 **Abstract**

26

27 Anastomosing branching and wavy laminae in quartz-rich sandstones of the 3.22 Ga Moodies
28 Group in the Barberton Greenstone Belt (BGB), South Africa, have been extensively described as
29 fossilized microbial mats developed in a terrestrial to marine transitional environment. Petrological
30 and geochemical characteristics of a ~350 m thick Moodies succession in the Saddleback Syncline
31 in the central BGB were analyzed to reconstruct the post-depositional history of the sediments and to
32 better constrain the origin of the laminae. The studied sandstones are composed mainly of quartz,

33 potassium feldspar, and chert clastic grains with various proportions of microquartz and sericite
34 cements. In coastal floodplain and inter- to supra-tidal environments, quiescent periods with low
35 current velocity allowed the repeated deposition of thin (<5 mm thick) and fine-grained laminae with
36 clay matrix and potentially organic matter. In contrast, subtidal settings under consistently high-
37 energy conditions led to the deposition of coarse- to medium-sized sands without fine-grained
38 lamina. Intergrain areas in those coarse sediments were infilled with a microquartz cement during
39 burial diagenesis. Due to the presence of clay matrix, the fine-grained laminae had a reduced
40 porosity that prevented the microquartz cementation within the laminae. This original clay matrix
41 was later replaced with sericite cements during metamorphism, forming the anastomosing and wavy
42 laminae observed at the hand-specimen scale in the studied sandstones. Microscopic dark areas
43 within the laminae are composed mostly of opaque minerals with scarce carbonaceous material. The
44 scarcity of carbonaceous material and the fluctuating energy settings, potentially inauspicious for the
45 continuous development and preservation of biomats, are not in favor of a flourishing Paleoproterozoic
46 microbial life on the Moodies sediments.

47

48 **1. Introduction**

49

50 Among the various clues to understand the record of traces of life on early Earth, ancient
51 microbial biofilms and microbially induced sedimentary structures (MISS) have been a subject in
52 numerous studies over the last 15 years (e.g., Noffke et al., 2003, 2008; Noffke, 2007; Wacey, 2009;
53 Hickman-Lewis et al., 2018; Lepot, 2020). The development of these structures depends largely on
54 the depositional setting through colonization of biofilms and their potential physical interactions
55 with clastic grains (Noffke et al., 2001). Some Archean rocks are composed of siliciclastic sediments
56 deposited in tidal and shallow shelf settings that may have been favorable to the development of
57 biofilms (e.g., Noffke, 2007, 2010; Noffke et al., 2013; Westall, 2008; Westall et al., 2011).
58 However, the preservation of Archean biofilms and/or MISS required specific and complex
59 taphonomic processes and their biogenicity has often been debated (e.g., Bower, 2011; Mariotti et
60 al., 2014; Davies et al., 2016, 2018; Noffke, 2018; Westall et al., 2018; Hickman-Lewis et al., 2019).

61 Noffke et al. (2006) first proposed that wrinkle and roll-up sedimentary structures in the ~3.22
62 Ga Moodies Group in the Barberton Greenstone Belt (BGB), South Africa, were MISS. Heubeck
63 (2009) subsequently interpreted that ubiquitous green anastomosing branching and wavy laminae in
64 the Moodies sandstones were remnants of microbial mats. Homann et al. (2015) classified the

65 laminae into three morphotypes and suggested a relationship between the mat morphologies and
66 their associated depositional environments. A systematic difference in carbon and nitrogen isotopic
67 composition between the terrestrial and marine deposits was also observed and interpreted to reflect
68 the oldest evidence for the co-existence of distinctive terrestrial and marine ecosystems (Homann et
69 al., 2018; Homann, 2019; Thomazo et al., 2020).

70 The BGB has undergone lower greenschist-facies regional metamorphism ($< 350^{\circ}\text{C}$) and has a
71 complex thermal history (e.g., de Ronde and de Wit, 1994), such as the emplacement of several
72 plutonic domains around the BGB at 3.23-3.21 Ga (Fig. 1a) (e.g., de Ronde and de Wit, 1994; Cutts
73 et al., 2014 ; Moyen et al., 2019), followed by multiple tectono-thermal events during the
74 Mesoarchean to Paleoproterozoic (Weis and Wasserburg, 1987; de Ronde et al., 1991a; Lécuyer et
75 al., 1994; Toulkeridis et al., 1994, 1998). The Moodies sediments in the BGB were presumably
76 affected by complex post-depositional processes, including polymetamorphism and more recent
77 alteration via fluid circulations (e.g., Heubeck, 2019; Bonnand et al., 2020); however, the influence
78 of post-depositional processes on the Moodies rocks has not yet been examined in detail.

79 In this study, we analyzed the microfacies and major and trace element composition of the
80 lamina-bearing Moodies sandstones. Based on petrological and geochemical results, we

81 reconstructed multiple post-depositional processes of the analyzed sandstones in association with
82 their depositional settings and with the regional metamorphic history of the BGB. They enabled us
83 to discuss the origin of the anastomosing and wavy laminae in the Moodies rocks in the framework
84 of their post-depositional history.

85

86 **2. Geological setting**

87

88 The BGB at the Makhonjwa Mountains is located in northeastern South Africa and northern
89 Eswatini (Swaziland) near the eastern margin of the Kaapvaal Craton (Fig. 1a). It is one of the best-
90 preserved Paleoproterozoic greenstone belts consisting of a >10 km thick succession of interlayered
91 volcanic and sedimentary rocks, termed the Barberton Supergroup (e.g., Lowe et al., 1999;
92 Hofmann, 2005). The Barberton Supergroup (ca. 3.55 to 3.22 Ga) is subdivided (in ascending order)
93 into the Onverwacht, the Fig Tree, and the Moodies groups. The Onverwacht Group (ca. 3.55 to 3.26
94 Ga) is composed mainly of mafic to ultramafic volcanic rocks (basalts and komatiites) with
95 interlayered cherts and rare felsic volcanics as well as very few clastic units. The lithologically
96 diverse Fig Tree Group (ca. 3.26 to 3.23 Ga) includes clastic sedimentary rocks (shales, sandstones

97 and conglomerates), immature volcanoclastics, intermediate to felsic lavas, banded iron formations,
98 barite, and cherts. The Moodies Group (ca. 3.22 Ga) is ~3.7 km thick and consists mainly of quartz-
99 rich sandstones and siltstones with subordinate conglomerates, felsic volcanics, shales and jaspilites
100 (Fig. 1b) (e.g., Eriksson, 1977, 1979, 1980; Heubeck and Lowe, 1994a, b, 1999; Homann et al.,
101 2015; Heubeck, 2019). The BGB is tightly folded; the central belt consists of a series of synclines,
102 mostly cored by Moodies Group strata and separated by strike faults replacing tight anticlines
103 (Anhaeusser et al., 1981; Heubeck and Lowe, 1994a, b).

104 The Moodies Group has been suggested to be the oldest terrestrial to marine transitional
105 sedimentary succession (e.g., Eriksson, 1977, 1979, 1980; Heubeck and Lowe, 1994a, b; Eriksson
106 and Simpson, 2000; Eriksson et al., 2006; Simpson et al., 2012; Homann et al., 2015; Heubeck,
107 2019). Heubeck et al. (2013) indicated that the entire Moodies Group accumulated very rapidly
108 (within <1–14 million years) at ~3.22 Ga, based on the dating of zircons in intercalated tuff beds of
109 the sedimentary succession. North of the Inyoka Fault, a major divide in the BGB, the Moodies
110 Group is exposed in several tectonic units, including (from northeast to southwest) the Eureka
111 Syncline, Dycedale Syncline, Saddleback Syncline, Moodies Hills Block, and Stolzberg Syncline
112 (e.g., Homann et al., 2015; Heubeck et al., 2016).

113 In the present study, we focused on a Moodies sedimentary succession in the Saddleback
114 Syncline in the central BGB (Fig. 1). The ~3000 m thick Moodies Group is exposed continuously
115 along the NE-SW trending syncline (Homann et al., 2015; Heubeck, 2019). In the Saddleback
116 Syncline, the bottom 50-80 m thick part of the Moodies Group is composed mainly of interbedded
117 cobble conglomerates and sandstones (Fig. 1b). These basal conglomerates are correlated to the
118 MdB unit of the group in the northern Eureka Syncline (Table 1) (Anhaeusser, 1976; Homann et al.,
119 2015), and are thought to have been deposited in a fluvial to alluvial setting (Eriksson, 1980;
120 Heubeck and Lowe, 1994a). This conglomerate unit is overlain by ~1000 m thick quartz-rich
121 sandstones in the lower part of the Saddleback succession. These sandstones are correlated to the
122 MdQ1 unit of the Eureka succession (Anhaeusser, 1976; Homann et al., 2015), and would have been
123 deposited on coastal floodplain to subtidal settings (Homann et al., 2015). Anastomosing branching
124 and wavy laminae are frequently observed in these sandstones and are interpreted to be remnants of
125 original microbial mats (Noffke et al., 2006; Heubeck, 2009; Homann et al., 2015).

126 The overlying 400-1000 m thick interval is composed mainly of fine-grained sandstones and
127 siltstones and is correlated to the MdS1 unit of the Moodies Group (Fig. 1b) (Anhaeusser, 1976).
128 This interval was deposited on a prodelta slope below the wave base (Heubeck, 2019). The MdS1

129 sandstones and siltstones are overlain by up to 150 m thick quartzarenites, which is correlated to the
130 MdQ2 unit in the Eureka Syncline (Anhaeusser, 1976). The MdQ2 unit is interpreted as reflecting
131 subtidal and intertidal settings (Eriksson, 1977), or an offshore subaqueous dune field (Heubeck and
132 Lowe, 1994a). The MdQ2 arenites are overlain by a basaltic lava (MdL2 in Anhaeusser, 1976). This
133 characteristic MdL2 lava is then overlain by banded iron formation (BIF), jaspilite, shale and
134 siltstone (MdI2 in Anhaeusser, 1976). The MdI2 unit is overlain by ~300 m thick sandstones with
135 conglomerates and siltstones, and these conglomerates and siltstones are correlated to the MdS2 unit
136 (Anhaeusser, 1976). The uppermost Moodies succession in the Saddleback Syncline is characterized
137 by up to 100 m thick quartzarenites and conglomerates (MdQ3).

138

139 **3. Analyzed sedimentary succession and its depositional settings**

140

141 In the present study, we analyzed a ~350 m thick succession in the upper part of the MdQ1
142 unit of the lower Moodies Group (25°50'19.9" S, 31°05'01.1" E) (Figs. 1c and 2a) (Anhaeusser,
143 1976; Homann et al., 2015). The MdQ1 unit is ~1000 m thick and is composed mostly of quartz-rich
144 sandstones. This MdQ1 unit corresponds to the MD1 unit of alluvial plain facies in Eriksson (1978).

145 Heubeck et al. (2013) interpreted that the MdQ1 sediments were deposited in a braided fluvial
146 system. Heubeck (2009) subdivided the MdQ1 unit into four subunits based on the lithofacies and
147 reconstructed their depositional settings: a coastal floodplain (subunit 1), a tidal coast (subunit 2),
148 and subtidal and shoreface settings (subunits 3 and 4) (Table S1). Gamper et al. (2011) substantially
149 changed the interpretation of their depositional settings: a terrestrial coastal zone (subunit 1), a
150 subtidal to intertidal zone (subunit 2), and a nearshore zone (subunits 3 and 4). Homann et al. (2015)
151 later revised these depositional settings based on macroscopic lithofacies description: a coastal
152 floodplain (subunit 1), an intertidal zone (subunit 2), a subtidal zone (subunits 3), and an upper inter-
153 to supra-tidal zone (subunit 4) (Fig. 1c). The analyzed succession in the present study includes the
154 four subunits of the MdQ1 unit and corresponds to stratigraphic Log 9 in Homann et al. (2015) in the
155 central Saddleback Syncline. In this study, we follow the interpretation of depositional setting of the
156 analyzed four subunits in Homann et al. (2015) (Figs. 1c and 2a).

157 The lowermost subunit 1 is ~200 m thick and is composed of coarse- to fine-grained
158 sandstones. Cross-bedding, herringbone structures, and channels with erosive base are observed on
159 the outcrop. Pebbles are most commonly observed in the lower part of the subunit (Fig. 2b).
160 Anastomosing branching and wavy laminations are frequently recognized at the hand-specimen

161 scale (Fig. 2c). Homann et al. (2015) interpreted the environment of the subunit 1 as a coastal
162 floodplain, based notably on low-angle planar and trough cross-bedding and lenticular
163 conglomerates with desiccation cracks.

164 The overlying subunit 2 is ~40 m thick and consists of medium-grained sandstones (Figs. 1c
165 and 2d). Abundant asymmetric ripples, common herringbone structures, and some small and local
166 pebbles are observed. Green anastomosing branching and wavy laminations at the hand-specimen
167 scale are recognized. Based on the presence of herringbone cross-bedding, sigmoidal foresets, and
168 mud drapes, Homann et al. (2015) suggested that the subunit 2 accumulated in an intertidal setting.

169 The ~80 m thick subunit 3 is composed mainly of medium- to fine-grained sandstones (Fig.
170 1c). Large cross-beddings are frequently observed through the subunit (Fig. 2e). As observed in
171 Homann et al. (2015), this subunit is characterized by the lack of anastomosing branching and wavy
172 laminations. The authors constrained its depositional environment to a subtidal setting, based on
173 large-scale and low-angle planar foresets and putative gutter casts.

174 The uppermost subunit 4 is ~40 m thick and consists of medium-grained sandstones (Fig. 1c).
175 Cross-beddings and subvertical gas-/fluid-escape structures are frequently observed (Figs. 2f and g).
176 Some anastomosing branching and wavy laminae are recognized. This subunit 4 corresponds to the

177 basal part of the MD3 unit in Eriksson (1979), in which this part was interpreted to be a delta-front
178 sediment. Homann et al. (2015) reinterpreted the depositional setting of the subunit 4 as an upper
179 inter- to supra-tidal setting, based on abundant desiccation cracks in the subunit. A slight difference
180 in thickness of the subunits 2 and 3 is observed between Log 9 in Homann et al. (2015) and the
181 present log (Fig. 1c). This could be explained by substantial lateral changes in thicknesses of
182 sedimentary bodies (megaripples and dunes) that form these subunits.

183

184 **4. Materials and analytical methods**

185

186 Forty-two rock samples were collected from the analyzed Saddleback succession. A part of
187 these samples has been previously analyzed for quadruple sulfur isotope geochemistry (Saitoh et al.,
188 2020). The lithofacies of all the collected rocks was observed in detail via optical microscopy. The
189 proportion of grain to matrix/cement in the rocks was estimated according to Baccelle and Bosellini
190 (1965). In addition to the microfacies description, multiple geochemical analyses (Raman
191 spectroscopy, EPMA, SEM-EDS, LIBS, ICP-OES, ICP-MS, and EA) were conducted on a selected
192 subset of the collected rock series.

193

194 4.1. Raman spectroscopy

195 Eight samples in the analyzed succession (SAD4, 6, 20, 21, 26, 34, 39, and 40) were analyzed

196 by Raman spectroscopy at the Laboratoire Magmas et Volcans (LMV), Clermont-Ferrand, France.

197 Raman spectra were collected using a Renishaw InVia confocal Raman micro-spectrometer

198 equipped with a 532 nm laser source, a Peltier-cooled CCD detector, an edge filter, a motorized

199 XYZ stage and a Leica DM 2500M optical microscope. Scattered light was collected by a back-

200 scattered geometry. Laser power on the sample was reduced by filters in order to operate at powers

201 <1 mW. We used a 1200 grooves/mm grating, which results in a spectral resolution better than 1

202 cm^{-1} . The slit aperture was set either to 20 μm (high confocality setting) or 65 μm (standard

203 confocality setting) depending on grain size. A 100 \times microscope objective was used. These

204 analytical conditions result in lateral and vertical spatial resolutions of <1 and 2 μm , respectively.

205 Daily calibration of the spectrometer was performed based on a Si 520.7 cm^{-1} peak. All spectra were

206 recorded in the 95-2695 cm^{-1} wavenumber range and from \sim 1050 to 3400 cm^{-1} in presence of

207 carbonaceous matter, using WIRETM 4.4 software. The number of acquisitions on a single spot

208 varied from three to fifteen, and the duration of each acquisition ranged between 5 and 30 s. Ninety-

209 eight spots were measured in total in the selected eight samples. 2D maps were acquired on several
210 areas using a 0.8 μm or a 2 μm step between points. Mineral identification was performed by
211 comparison with standard spectra from the WIRE and RRUFF databases.

212

213 4.2. Electron Probe Micro Analyzer (EPMA)

214 Selected nine samples were analyzed by EPMA at the University of Lausanne, Switzerland.
215 Quantitative spot analyses for Si, Mg, Na, K, Al, Ti, Fe, Ca, and Cr were conducted for five samples
216 (SAD5, 22, 32, 37, and 39) with a JEOL 8230F microprobe, using a beam current of 400 nA and an
217 accelerating voltage of 15 kV, with a dwell time of 100 ms. Lab standard minerals (albite for Si and
218 Na, forsterite for Mg, orthoclase for K and Al, TiO_2 for Ti, fayalite for Fe, anorthite for Ca, and
219 chromite for Cr) were used for spot analyses. Moreover, X-ray compositional maps for Si, Mg, Na,
220 K, Al, Ti, and Fe were obtained for five samples (SAD20, 23, 26, 34, and 39) with the same
221 analytical conditions.

222

223 4.3. Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS)

224

225 SEM-EDS measurements were conducted for five samples in the subunits 1 and 2 at the
226 University of Lausanne, using a Tescan Mira LMU SEM equipped with the Oxford Instruments
227 AZtec 3.4 software package, to identify mineral phases in the sandstones. The measurements were
228 operated at 20 kV acceleration voltage, ~6 pA probe current, and 21 mm working distance.
229 Backscattered electron (BSE) images were taken on finely polished thin sections coated with a 15
230 nm carbon layer, and the EDS analyses were carried out on them to determine the chemical
231 composition of minerals in the analyzed areas. We conducted both spot analyses and semi
232 quantitative elemental mapping, and the spatial distribution of Si, K, Al, Na, Ti, Fe, and Mg were
233 visualized in the maps.

234

235 4.4. Laser-induced breakdown spectroscopy (LIBS) imaging

236

237 LIBS imaging analyses were conducted for one sample from the subunit 2 at the Institute of
238 Light and Matter, Lyon, France. In LIBS imaging, a series of laser-induced plasma is generated at
239 different positions on the sample. Such plasma sources allow specific optical responses resulting
240 from the electronic relaxation of atoms and ions excited by the high plasma temperature to be

241 elicited from the elements constituting the sample. The light emitted by the plasma is collected and
242 analyzed using an optical spectrometer equipped with an intensified charge-coupled device (ICCD)
243 camera. The elemental “signal” (atomic and ionic emissions) is then extracted from the recorded
244 spectra, and elemental maps can be obtained in a pixel-by-pixel manner (Sancey et al., 2014; Fabre
245 et al., 2018; Motto-Ros et al., 2020). The sandstone sample was analyzed using Nd:YAG laser with
246 pulse energy of about 600 μ J operating at 100 Hz, and a lateral resolution (i.e. distance between two
247 consecutive laser shots) of 25 μ m. Two different Czerny-Turner spectrometers were used in order to
248 detect intense lines of Al (309.2 nm), B (208.9 nm), Cr (267.7 nm), Fe (302.0 nm) and Ti (323.4
249 nm), among others.

250

251 4.5. Major and trace element geochemistry

252

253 Fresh parts of the rock samples were carefully selected for major and trace element
254 measurements and cut to remove weathered surfaces. The rock pieces were crushed into small chips
255 (<1 cm in diameter) and were ultrasonically cleaned with distilled water and subsequently with pure
256 acetone. The cleaned chips were then powdered using an agate mill. Up to 25 g of powder was

257 prepared for each sample. Twenty-nine samples from the four subunits were selected for major
258 element analyses at the LMV in Clermont-Ferrand. Sample powder (100 mg) was mixed with LiBO₂
259 flux (300 mg) and fused at 1100 °C. The solutions were analyzed using the LMV HORIBA-Jobin-
260 Yvon ULTIMA-C Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). The
261 precision and accuracy were determined from repeated measurements of the BHVO-1 reference
262 material. Results are consistent within 3% of the certified data except for K₂O (4%). The total
263 volatile content (loss on ignition) was determined from 1g of sample powder.

264 Trace elements were measured on twelve samples from the four subunits at the LMV. About 50
265 mg of sample powder were digested using ammonium bifluoride (Zhang et al., 2012). Closed
266 savillex Teflon vials were placed into an oven at 220°C for 24 hours. After cooling, samples were
267 dissolved in concentrated HNO₃ and evaporated to dryness several times. All measurements were
268 performed using the LMV Agilent Inductively Coupled Plasma Mass Spectrometer (ICP-MS) in
269 Clermont-Ferrand, France. Samples and certified rock standard were measured in 0.5M-HNO₃–
270 0.05M-HF with a dilution factor of about 3,500. Counts per second were calibrated using certified
271 rock standards BHVO-2. The blank and reference materials (BE-N, BIR-1 and BHVO-2) were
272 measured every three samples. Measurements were corrected from oxide interferences and

273 instrumental drift if significant. The precision and accuracy were determined from four repeated
274 measurements of the BE-N and BIR-1. Results are consistent within 4% of published data, except
275 for U, Th, and Pb (<10%).

276 Trace element concentrations of the analyzed sandstones were normalized to upper continental
277 crust (UCC) composition (Rudnick and Gao, 2014), and their rare-earth element (REE)
278 concentrations were normalized to Chondrite and to Mud from Queensland (MuQ) (McDonough
279 and Sun, 1995; Kamber et al., 2005).

280

281 4.6. Elemental Analyzer (EA)

282

283 For three samples from the subunits 1, 3, and 4, total organic carbon (TOC) contents were
284 analyzed by EA. Powdered samples (~0.5 g) were reacted with 6N HCl at room temperature for >12
285 h to remove carbonate at the LMV. The residue was cleaned by repeated centrifugation with
286 ultrapure water and was then reacted with 46M HF at 55°C for >48 h to dissolve silicate. The
287 residue was cleaned by repeated centrifugation with ultrapure water and was dried at 55°C for >12 h.
288 Aliquots of the dried residue (1–20 mg) were weighed in a tin capsule and their TOC contents were

289 measured by a vario MICRO cube elemental analyzer (Elementar GmbH, Hanau, Germany) at
290 Laboratoire Biogéosciences, Université Bourgogne Franche-Comté, Dijon, France. USGS40-
291 certified reference material (C = 40.8 wt.%) was used for calibration and the TOC content is shown
292 as a dry weight percentage of the total fraction. The analytical reproducibility, based on duplicate
293 analyses of USGS40, is better than 0.1 wt.% (1σ) for the residue.

294

295 **5. Results**

296

297 5.1. Petrological description

298

299 *5.1.1. Clastic grains*

300 The analyzed Moodies succession is composed mostly of sandstones with some conglomerates
301 (Figs. 1-3). The sandstones contain abundant coarse to very fine sand-size clastic grains. The grains
302 range in size mostly from 100 to 1000 μm . Those clastic grains are mainly quartz (>50%) (Figs. 3a
303 and 3b). Potassium (K) feldspar (microcline and orthoclase) (<25%) and chert grains (<15%) are
304 also frequently observed and the latter consists mostly of microquartz crystals with intracrystalline

305 fine sericite (Figs. 3c and d). As some chert grains include substantial amounts (>5%) of sericite,
306 Heubeck and Lowe (1999) described them originally as “quartz-sericite (QS) grains”. A small
307 amount of albite (<2%) is also contained in the sandstones. The roundness and sphericity of quartz
308 and K-feldspar grains are variable but larger grains (>400 μm in diameter) show lower roundness
309 and sphericity (<0.3 and <0.4, respectively). In contrast, the roundness and sphericity of chert grains
310 are relatively higher (mostly >0.7 and >0.7, respectively). The sandstones are poorly sorted through
311 the analyzed succession.

312 Many clastic grains show long to concavo-convex intergranular pressure solution (IPS)
313 textures (arrows in Fig. 3d). Undulose extinction is often recognized in monocrystalline quartz
314 grains (Fig. 3b). Some quartz grains contain apatite ($\text{Ca}_5(\text{PO}_4)_3\text{Cl}$) crystals as inclusion (Figs. 3e and
315 g). These apatite inclusions are monocrystalline and elongated (length close to 30 μm). Some K-
316 feldspar grains are altered patchily by albite ($\text{NaAlSi}_3\text{O}_8$) (Supplementary Fig. S1). Rounded detrital
317 zircon grains (<200 μm in diameter) are present in the sandstones. The analyzed sandstones also
318 contain small monazite-(Ce) grains (Figs. 3f and h). The grain size of the monazite is mostly <10
319 μm , and its particle shape is generally irregular. Monazite grains sometimes aggregate within the
320 intergrain areas. An overgrowth texture is not recognized within a monazite crystal. Our description

321 of the Moodies clastic grains is generally consistent with that in previous studies (Heubeck and
322 Lowe, 1999; Hessler and Lowe, 2006; Heubeck, 2019), in which quartz (70-90%) and K-feldspar
323 (<20%) are major clastic grains in the MdQ1 sandstones in the eastern Saddleback Syncline.

324

325 *5.1.2. Intergrain areas*

326 Most of the intergrain areas are filled with microquartz and sericite ($\text{KAl}_2\text{AlSi}_3\text{O}_{10}(\text{OH})_2$) (Fig.
327 4), although micropores (mostly <200 μm in diameter) are also observed in the sandstones
328 throughout the succession. These microquartz and sericite were classified originally into
329 “pseudomatrix” in Heubeck and Lowe (1999). However, we describe them as a cement in this study
330 because their fabrics suggest that they recrystallized from an original matrix (see below and Sections
331 6 and 7). No original clay matrix is identified clearly in the present sandstones. This observation is
332 consistent with the previous interpretation that the original matrix in the Moodies sandstones has
333 been recrystallized totally to a mosaic of fine-grained sericite and microquartz (Heubeck and Lowe,
334 1999). The microquartz cements fill irregular-shaped spaces between the clastic grains. The sericite
335 cements are also observed around the detrital grains. The sericite crystals are normally elongated up
336 to 30 μm (Fig. 4d). Muscovite is also recognized in the sandstones commonly filling spaces between

337 the clastic grains (Fig. 3e). The size of muscovite crystals is up to 200 μm and their shapes are
338 usually irregular but sometimes subhedral. Some veins filled with sericite cut the microquartz
339 cements and clastic grains (Figs. 4e and f).

340 Through the analyzed succession, the microquartz cements are generally dominant in the
341 coarser sandstones. In contrast, fine-grained laminae in the subunits 1, 2 and 4 (Fig. 1c), which are
342 frequently intercalated into the coarse sediments, are enriched mostly in sericite (Figs. 4b and c).
343 The subunit 3 lacks such fine-grained laminae and thus the intergrain areas are filled mainly with
344 microquartz cements (Fig. 4a). In the subunits 1 and 2, the relative proportion of clastic grains to the
345 microquartz and sericite cements is mostly between 40 and 50%. The proportion is higher in the
346 overlying subunit 3 (up to 60%), reflecting the lack of sericite-bearing fine-grained lamina in this
347 subunit. In the uppermost subunit 4, the proportion is between 35 and 50%.

348

349 *5.1.3. Accessory minerals*

350 Various types of oxide mineral are observed in the analyzed sandstones (Fig. 5). Titanium (Ti)
351 oxide particles are common throughout the succession (Fig. 5a). Ti oxides are sometimes observed
352 as aggregates of small (<20 μm) particles. These aggregates are mostly irregular-shaped and

353 sometimes coat clastic grains or fill intergrain areas (Fig. 5b). The Raman results show that the Ti
354 oxides are rutile and anatase. Some veins bearing Ti oxides cut both of the microquartz and sericite
355 cements and clastic grains (Figs. 5c-f and 7). Iron (Fe) oxide grains are also recognized in the
356 sandstones. The Fe oxides are irregular shaped and their sizes are mostly <50 μm . The Fe oxides are
357 observed within the cements along grain boundaries. Some Fe oxides are also observed along a
358 micropore (Fig. 5g). The Raman results show that some Fe oxides are goethite. The macroscopic
359 LIBS images demonstrate that Fe is mainly concentrated in the secondary altered parts of the
360 sandstones (Supplementary Figs. S2 and S3). The common occurrence of accessory Fe-oxides in the
361 Moodies sandstones is consistent with the previous description in Heubeck and Lowe (1999). Small
362 sulfide grains are also contained in the analyzed sandstones (Fig. 5h) (Saitoh et al., 2020). The
363 sulfides are exclusively pyrite and range in size mostly from 5 to 100 μm . The shape of the pyrite
364 crystals is mainly irregular but some are euhedral to subhedral. The pyrite crystals are mostly within
365 the cements and along grain boundaries.

366

367 *5.1.4. Anastomosing and wavy laminae*

368 On the outcrop, anastomosing branching and wavy laminae are frequently observed in the

369 subunits 1, 2 and 4 of the analyzed succession (Figs. 1c, 2c, and 6) (also see log N°9 in Homann et
370 al., 2015). The laminae are dark greenish colored at the hand-specimen scale when observed with the
371 naked eye and are continuous laterally over several to tens of meters. Microscopic observations
372 show that the laminae at the hand-specimen scale correspond to thin (mostly <5 mm thick) and fine-
373 grained laminae intercalated into coarse-grained sandstones (Figs. 2c and 6). Homann et al. (2015)
374 classified these laminae into three morphotypes. They suggested that planar-type laminae are a
375 characteristic of coastal floodplain (subunit 1), whereas wavy-types were typically formed in an
376 intertidal setting (subunit 2). Tufted-types characterized upper inter- to supra-tidal settings (subunit
377 4). However, we observed frequent planar- and wavy-type laminae in the subunits 1, 2, and 4, and no
378 tufted-type lamina was observed in the subunit 4. The subunit 3 is characterized by the lack of the
379 anastomosing branching and wavy lamina, as previously observed in Gamper et al. (2011) and
380 Homann et al. (2015).

381 The laminae are composed of finely fragmented quartz and K-feldspar grains (<250 µm in
382 diameter) (Figs. 6 and 7). The intergrain areas in these laminae are composed dominantly of sericite
383 with minor microquartz cements (Figs. 6c-f). Opaque minerals are commonly observed within the
384 laminae and are composed mostly of rutile and anatase and more occasionally of goethite (Fig. 8).

385 Some Ti oxides show an anastomosing microtexture (Figs. 8a and d). In some cases, Ti oxide
386 particles are observed at the boundary between the fine-grained lamina and coarse-grained sediments
387 (Fig. 6e). The Ti concentration within the laminae is confirmed by the EPMA (Fig. 7) and LIBS
388 (Supplementary Figs. S2) images. The LIBS images also show the presence of chromium and boron
389 within the laminae, which are likely incorporated into sericite.

390 Carbonaceous material occurs scarcely within the anastomosing and wavy laminae in the
391 subunits 1, 2, and 4 (Fig. 1c), as shown by Raman analyses (Figs. 8 and 9). The carbonaceous
392 material was rarely detected, even in laminae, for the seven samples investigated. Only some
393 micrometric regions in one lamina in SAD39 were enriched in carbonaceous material (Fig. 9). Note
394 that the Raman maps are visual representations of a binary matrix, i.e., colored pixels rely on the
395 presence of a given organic/mineral phase, but do not inform on its concentration. In accordance
396 with the previous Raman data in Homann et al. (2018), the detected carbonaceous materials show a
397 Raman signature typical of disordered carbonaceous material that has been submitted to greenschist-
398 facies metamorphism, with an intense and narrow D1 band, a less intense G band, and a D2 band
399 appearing as a shoulder (Figs. 8c and 9f). In the lamina in SAD39, carbonaceous materials occur
400 commonly in association with Ti oxide (rutile and anatase) (Figs. 9c-e): both of carbonaceous

401 material and Ti oxide are frequently detected at the same spot (Fig. 9f).

402

403 5.2. Major and trace element geochemistry

404

405 Major element diagrams of the analyzed Moodies sandstones are shown in Fig. 10 and
406 Supplementary Fig. S4. All the data are reported in Table 2. The results demonstrate that Si, Al, and
407 K are the three major elements of the analyzed sediments. This is consistent with the microscopic
408 observation that quartz, K-feldspar, and chert clastic grains and microquartz and sericite cements are
409 the major components of the rocks. The SiO₂ contents of the analyzed rocks are mostly between 75
410 and 95 wt.%. The SiO₂ contents of the subunit 3 are higher (>85 wt.%) than of other subunits (Fig.
411 1c), which is consistent with the higher proportion of quartz cement in the intergrain area and the
412 lack of laminae with terrigenous matrix in the subunit (Fig. 10a). Other than that, no major chemical
413 difference is observed in the different subunits. The Na₂O content of the sample (SAD18) in the
414 subunit 1 is substantially high (> 5 wt.%, see Table 2) and it is consistent with its extensive
415 albitization observed under microscope. The Al₂O₃ contents of the sandstones are correlated with the
416 SiO₂ ($R^2=0.97$), TiO₂ ($R^2=0.75$), K₂O ($R^2=0.48$), and Fe₂O₃ contents ($R^2=0.44$) (Figs. 9a and b and

417 Supplementary Fig. S5) (Table 2). All the subunits share similarly low loss of ignition (LOI)
418 between 0.47 and 2.55 wt.% (Fig. 10d). The major element composition of the analyzed sandstones
419 is similar to that of previously analyzed Moodies sandstones (orange circles in Fig. 10). In particular,
420 the present sandstones share the low Fe₂O₃, MnO, and MgO contents with the only one sandstone
421 sample from the Saddleback Syncline available in the literature (orange dashed arrows in Fig. 10)
422 (Hessler and Lowe, 2006).

423 The trace element concentration of the analyzed Saddleback sandstones is reported in Table 3,
424 and their patterns normalized to the upper continental crust (UCC) are shown in Supplementary Fig.
425 S6. Chondrite- and MuQ-normalized REE patterns are shown in Fig. 11. The trace element patterns
426 of the analyzed sandstones are similar to the modern UCC pattern and show slight depletion in Cs,
427 Nb, Ta, Pb, Sr, V, Co, and Zn and enrichment in Li, Sc, and Cr. The MuQ-normalized patterns show
428 no positive Eu anomaly ($\text{Eu}/\text{Eu}^* < 1.12$). No clear difference is observed between the different
429 subunits in the analyzed succession. The present trace element data are consistent with the previous
430 data of the Moodies sandstones (Hessler and Lowe, 2006).

431

432 **6. Chronology of depositional and post-depositional processes**

433

434 Based on the petrological descriptions and the geochemical results, we propose a scenario of
435 several processes that have affected the studied Moodies sediments sequentially during and after
436 their deposition (Fig. 12).

437

438 6.1. Stage 1: Sedimentation

439

440 The first stage corresponds to the deposition of detrital grains, mainly quartz, K-feldspars, and
441 chert fragments (Fig. 12a). Quartz and feldspar grains derived mostly from plutonic and felsic
442 volcanic rocks (e.g., Heubeck and Lowe, 1999; Hessler and Lowe, 2006), whereas chert grains were
443 presumably derived from older sedimentary rocks of the Onverwacht and Fig Tree groups (e.g.,
444 Heubeck and Lowe, 1999; Saitoh et al., 2020).

445 The analyzed sediments are subdivided into two types according to their depositional settings.

446 The sediments of the subunit 3 deposited in a subtidal environment (Fig. 1c), in which a constant and
447 high current velocity led to the deposition of coarse grains through large sand bodies (dunes and
448 megaripples, also see Homann et al., 2015). In such a high energy sandy setting, most of fine-

449 grained clay and potential organic matter may have been winnowed and not have accumulated (e.g.,
450 Noffke et al., 1997). Accordingly, the SiO₂ and Al₂O₃ contents of samples from the subunit 3 are
451 higher and lower, respectively, than those from other subunits (Fig. 10a). In marked contrast, the
452 sediments of the subunits 1, 2, and 4 deposited in coastal floodplain and inter- to supra-tidal settings
453 (Fig. 1c) (Homann et al., 2015). These depositional settings were likely characterized by periodical
454 changes in current velocity under the influence of river flush or tides. When the current velocity was
455 temporarily low, relatively fine detrital grains accumulated possibly with a matrix (clay minerals and
456 organic matter), contributing to the formation of a fine-grained lamina intercalated into coarser
457 sediments (Fig. 12a). Although such original clay matrix is not petrographically observed in the
458 present rocks (Figs. 4 and 6), It is commonly recognized in other Moodies localities (e.g., Eriksson
459 and Simpson, 2000; Homann et al., 2018).

460 In the present sandstones, the supposed presence of the original clay matrix is consistent with
461 the major element geochemistry of the rocks. The Al₂O₃ contents are well-correlated with the SiO₂,
462 K₂O, TiO₂, and Fe₂O₃ contents (Figs. 10a and b and Supplementary Fig. S5), along a trend with
463 other Moodies shales and sandstones (McLennan and Taylor, 1983; Hessler and Lowe, 2006;
464 Toulkeridis et al., 2015), and igneous clasts in Moodies conglomerates (Sanchez-Garrido et al.,

465 2011; Agangi et al., 2018). These correlations suggest that the major element composition of the
466 initial Moodies sediments was controlled mainly by the relative proportion of quartz and clay matrix,
467 because a similar correlation is commonly observed in modern terrigenous sediments and typically
468 reflects the effect of hydrodynamic sorting of Al₂O₃-, K₂O-, Fe₂O₃-, and TiO₂-rich clay from coarse
469 quartz particles in distal and proximal environments (e.g., Singh, 2009). In the subunits 1, 2, and 4 of
470 the present Saddleback succession, more quiescent periods leading to the deposition of fine grains
471 may also have been favorable for potential biofilm installation (e.g., Noffke et al., 1997; Homann et
472 al., 2015).

473

474 6.2. Stage 2: Microquartz cementation

475

476 After the initial sedimentation phase, an interstitial fluid enriched in silica led to microquartz
477 precipitation and cementation between detrital grains in the analyzed Moodies sediments (Fig. 12b).
478 Three potential processes can be considered for the microquartz cementation: 1) low-temperature
479 sediment-water interaction at or below the surface (e.g., Knauth and Lowe, 1978; Lowe, 1999), 2)
480 microquartz precipitation from a hydrothermal fluid (e.g., Knauth and Lowe, 1978; de Wit et al.,

481 1982; Hofmann, 2005; Van Kranendonk, 2006), and 3) IPS and subsequent microquartz cementation
482 during burial (e.g., McBride, 1989; Tada and Siever, 1989).

483 Our observation suggests that IPS was the main mechanism for the microquartz cementation in
484 the analyzed sediments, for the following reasons. Firstly, the gas-/fluid-escape structures are
485 frequently observed in the analyzed Moodies sandstones (Figs. 2f and g). These structures should
486 have formed in an early diagenetic stage before sediment consolidation (e.g., Lowe, 1975; Homann
487 et al., 2015). The well-preserved and silicified gas-/fluid-escape structures suggest that the
488 microquartz cementation and sediment silicification occurred during burial and are not concomitant
489 with sedimentation (Heubeck, 2019). Microquartz precipitation around the sediment-water interface
490 is therefore unlikely. Secondly, the trace element patterns are similar in all the subunits and show no
491 substantial depletion in mobile elements (Supplementary Fig. S6), implying that the microquartz
492 cementation from an additional hydrothermal fluid was not significant.

493 Finally, the frequent IPS textures in the analyzed sandstones (Fig. 3d) suggest that the pressure
494 solution during burial contributed substantially to the microquartz cementation. The microquartz
495 cement is commonly observed in the coarser sediments throughout the analyzed succession. It is
496 likely that these coarse sediments were characterized by high intergrain porosity during their

497 deposition in a relatively high-energy setting (Stage 1), and were favorable to IPS and silicification
498 during burial diagenesis. The high SiO₂ contents of coarse sandstones in the subunit 3 (>85 wt.%;
499 Fig. 10a) may partly reflect the intense post-depositional silicification, because mineral sorting alone
500 rarely leads to such substantially high SiO₂ contents (e.g., Singh, 2009). In contrast, the microquartz
501 cement is less present within the fine-grained laminae in the subunits 1, 2, and 4 (Figs. 1c, 4b and c).
502 The original clay matrix with low porosity likely prevented pervasive microquartz cementation
503 within the laminae.

504

505 6.3. Stage 3: Sericite cementation

506

507 The sericite cements are frequently observed in the analyzed Moodies sediments and are
508 particularly common within the fine-grained laminae in the subunits 1, 2, and 4 (Figs. 1c, 4, and 6c-
509 f). Some sericite veins clearly cut micro-quartz cements (Figs. 4e and f). This geometric relationship
510 implies that the sericite cementation occurred after the sediment silicification and consolidation
511 (Stage 2). We interpret that the original clay matrix within the fine-grained laminae (Stage 1) was
512 replaced with sericite by a fluid circulation along the bedding during later metasomatism (Fig. 12c).

513 The major and trace element geochemistry of the analyzed Moodies sandstones does not show
514 a typical feature of sediments being flushed at high temperature with high water/rock ratios (Figs. 10
515 and 11 and Supplementary Fig. S6) (e.g., bell-shape in the REE pattern and depletion in moderately
516 immobile elements; McLennan, 1989). This suggests that the sericite cementation was not due to a
517 hydrothermal fluid circulation. Rather, the major and trace element composition of the analyzed
518 sandstones is similar to that of their potential sources (i.e., igneous clasts in the Moodies; Agangi et
519 al., 2018). It indicates that secondary processes were not intense enough to leach out less-mobile
520 elements and to erase the geochemical information about the provenance of the sediments (Hessler
521 and Lowe, 2006). The analyzed Moodies sandstones have not been altered substantially by a
522 hydrothermal fluid. We thus infer that the sericite cementation occurred during metasomatism, with
523 a relatively low-temperature fluid, that has been commonly observed in the entire BGB rocks (e.g.,
524 Condie et al., 1977; de Ronde and de Wit, 1994; Hofmann, 2005), including the Moodies Group
525 (e.g., Heubeck and Lowe, 1999; Heubeck, 2019). The present geochemical data suggest that the
526 original clay matrix in the laminae was replaced with sericite during low-grade and fluid mediated
527 metamorphism (Dunoyer de Segonzac, 1970; Heubeck 2019).

528

529 6.4. Stage 4: Ti oxide formation

530

531 Ti oxides (rutile and anatase) are common in the analyzed sandstones (Fig. 5a). They form
532 aggregates of small (<20 μm) particles, coat clastic grains, fill irregular-shaped intergrain areas, and
533 represent anastomosing textures (Figs. 5b and 8). The dark and microscopic Ti-oxide laminae follow
534 parallel to the bedding and are present mainly in the fine-grained sericite-rich laminae. Also, some
535 veins bearing Ti oxides cut both of clastic grains and microquartz and sericite cements (Figs. 5c-f).
536 All these textures indicate that the Ti oxides formed secondarily after the microquartz and sericite
537 cementation (Stage 2 and Stage 3, respectively) (Fig. 12d). The Ti oxides can commonly form
538 during diagenesis and low-grade metamorphism via recrystallization of Ti-bearing minerals like
539 ilmenite, biotite and feldspars (e.g., Richards et al., 1988; Smith et al., 2009; Bower, 2011; Galvez et
540 al. 2012). Ti is highly stable and insoluble in a fluid (e.g., Brookins, 1988), though it can be more
541 mobile in the presence of phosphate ions (Pe-Piper et al., 2011). Our observations suggest that the Ti
542 oxides formed via the reaction of Ti-bearing detrital grains during metamorphism (Figs. 12a and d).

543 A close association of carbonaceous material and Ti oxide in sedimentary rocks has been
544 reported (Foucher et al., 2012; Galvez et al., 2012; Lekele Baghekema et al., 2017; Sirantoine et al.,

545 2020), even in fossilized microbial mats (Noffke et al., 2008). Although the carbonaceous material
546 has been observed only in the laminae that contain Ti oxides in the present sandstones (Figs. 8 and
547 9), the general scarcity of carbonaceous material through the analyzed succession does not allow us
548 to conclude firmly on the systematic association of carbonaceous material with Ti oxide. The origin
549 of this apparent association is still unknown and various processes have been suggested for it from
550 biological accumulation to post-depositional and diagenetic mobilization (Glamoclija et al., 2009;
551 Sirantoine et al., 2020).

552

553 6.5. Stage 5: Rainwater infiltration

554

555 Finally, Fe oxides precipitated during a recent oxic rainwater infiltration into the analyzed
556 Moodies sediments (Fig. 12e) (Heubeck, 2019). The Fe oxides are irregular shaped and observed
557 within the cements along grain boundaries and rarely within a fine-grained lamina. Some Fe oxides
558 are observed along a micropore (Fig. 5g). The occurrence of Fe oxide along a micropore is
559 consistent with the rainwater infiltration scenario into the sediments. The macroscopic LIBS images
560 also suggest that Fe is concentrated mainly in the reddish-brown superficially altered parts and that

561 its distribution is unrelated to the bedding in the analyzed sandstones (Supplementary Figs. S2 and
562 S3). A plausible Fe source for the oxides is pyrite crystals in the sandstones (Saitoh et al., 2020).
563 Although the timing of the rainwater infiltration and Fe oxide formation is poorly constrained,
564 Heubeck (2019) suggested that they occurred very recently. It is apparently consistent with the work
565 by Bonnard et al. (2020), showing that the emplacement of the negative Ce anomalies of the
566 Moodies BIF samples occurred in the last 100 Ma.

567

568 6.6. Correspondence to the regional thermal history

569

570 During the Moodies deposition, the emplacement of several plutonic domains occurred around
571 the BGB at 3.23-3.21 Ga (Fig. 1a) (e.g., de Ronde and de Wit, 1994), including the Kaap Valley
572 Pluton at ~3.21 Ga (e.g., Moyen et al., 2019 and references therein) and the Archean Gneiss
573 Complex (AGC) at 3.24-3.07 Ga (e.g., Taylor et al., 2012; Kröner et al., 2018; Wang et al., 2020),
574 associated with the major D2 deformation event in the BGB (e.g., de Ronde and de Wit, 1994). The
575 3.23-3.21 Ga emplacement was followed by major tectono-thermal events at ~3.1, ~2.7, and ~2.1 Ga
576 (Weis and Wasserburg, 1987; de Ronde et al., 1991a; Lécuyer et al., 1994; Toulkeridis et al., 1994,

1998). The extensive emplacement of the large plutons of the granodiorite-monzogranite-syenite (GMS) occurred at ~3.1 Ga (beige colored areas in Fig. 1a) (e.g., Anhaeusser et al., 1981). The wide distribution of this event indicates that the BGB sediments, including the Moodies, were substantially affected by associated heating. There are a number of gold deposits in the northern central BGB, some in close proximity (<10 km) to the analyzed Saddleback section (Munyai et al., 2011; Selvaraja et al., 2017). These gold deposits formed at ~3.1-3.0 Ga based on rutile and titanite U-Pb ages (de Ronde et al., 1991a, 1992; Agangi et al., 2019), presumably in association with the GMS event. This age is identical to the Sm-Nd age of carbonate-derived clay minerals in the Fig Tree Group in the central BGB (Toulkeridis et al., 1994), supporting the large influence of the GMS emplacement on the BGB sediments. Another major thermal event occurred widely in the BGB at ~2.7 Ga (e.g., de Ronde et al., 1991b; Toulkeridis et al., 1998). Toulkeridis et al. (1998) suggested extensive silicification and sericitization of carbonates in the Onverwacht and Fig Tree groups by low-temperature fluids (~200°C) at ~2.7 Ga, resetting the Pb-Pb, Sm-Nd, and Rb-Sr isotopic systems. The ~2.7 Ga thermal event was followed by a later one widespread in the BGB at ~2.1 Ga, based on Rb-Sr, Sm-Nd, and ⁴⁰Ar/³⁹Ar dating of the Onverwacht and Fig Tree sediments (e.g., Weis and Wasserburg, 1987, de Ronde et al., 1991b).

593 The present Moodies sandstones in the central BGB were presumably affected by all these
594 polymetamorphism/tectono-thermal events (e.g., Heubeck and Lowe, 1999). Previously published
595 Raman temperature estimates on Moodies carbonaceous materials provided temperature ranging
596 between 350 and 375°C (Nabhan et al. 2017; Homann et al. 2018; Kohler and Heubeck, 2019). If the
597 microquartz cements formed during burial diagenesis, the sericite cementation (Stage 3; Fig. 12c)
598 and Ti oxide formation (Stage 4; Fig. 12d) in the analyzed Moodies sandstones could correspond to
599 any of the multiple tectono-thermal events that occurred at a regional scale between ~3.1 and ~2.1
600 Ga.

601

602 **7. Origin of the analyzed Moodies anastomosing and wavy laminae**

603

604 Noffke et al. (2006) were the first to propose the presence of various types of MISS (e.g.,
605 wrinkle and roll-up structures) in the Moodies sediments from the Dycedale and Saddleback
606 synclines. Later studies generalized the presence of fossilized microbial mats in the Moodies
607 sediments as anastomosing and wavy laminae (Heubeck, 2009, 2019; Homann et al., 2015, 2018;
608 Homann, 2019; Köhler and Heubeck, 2019). Specific ecological and taphonomic conditions are

609 generally required for the formation and preservation of microbial mats in ancient sandstones
610 (Noffke et al., 2001). Along the different Moodies successions in the Saddleback Syncline, Homann
611 et al. (2015) argued in favor of morphological adaptation of microbial mats (planar-, wavy-, and
612 tufted-types) to the different paleoenvironments (coastal, intertidal, and inter- to supratidal settings,
613 respectively). Unfortunately, we have not been able to reproduce such a correlation between mat
614 morphology and depositional setting along the studied section. The identification of structures
615 morphologically similar to microbial mats in Moodies fluvial conglomerates also led some
616 authors to conclude the existence of a Paleoproterozoic terrestrial biosphere (Homann et al., 2018). The
617 presence and preservation of biofilms or anastomosing and wavy laminae in the Moodies sediments
618 thus seem to occur in various depositional settings (Homann, 2019).

619 Some Moodies environments are nevertheless characterized by the absence of such laminae as
620 seen in the subunit 3 of the Saddleback Syncline succession. The sediments of this subunit
621 accumulated in a relatively shallow subtidal setting, characterized by strong tidal currents and storms
622 (Homann et al., 2015). Under such consistently high-energy conditions, putative biofilms or a matrix
623 with organic matter would be likely reworked and vanished (Gerdes et al., 2000; Noffke et al.,
624 2003). In contrast, coastal floodplain and supra- to inter-tidal settings under fluctuating energy

625 conditions, corresponding to the present subunits 1, 2, and 4, are known to be more suitable for the
626 biomat formation and preservation (e.g., Noffke et al., 1997; Noffke, 2010; Newman et al., 2017). In
627 these latter subunits, the laminae are relatively continuous laterally over several to tens of meters and
628 are present stratigraphically almost every centimeter over a >200-m-thick succession. Archean to
629 modern examples of MISS that developed in tidal environments indicate that, even when microbial
630 biofilms can tolerate high energy events, their installation and development require a period of no
631 sediment accumulation under quiet conditions (e.g., Gerdes and Klenke, 2007; Eriksson and Wilde,
632 2010; Noffke, 2010; Cuadrado et al., 2013). Such ecologic and taphonomic windows of microbial
633 mat development and preservation (also see Noffke, 2009) appear questionable in the light of the
634 rather tight and continuous record of anastomosing and wavy laminae, with little evidence for
635 tearing and transportation of biofilm fragment, along the analyzed Moodies succession.

636 Based on the present results, we reinterpret the formation of the anastomosing and wavy
637 laminae observed along the studied succession in the Saddleback Syncline, previously suggested to
638 be ancient microbial mats (Homann et al., 2015) (Fig. 12). Our petrological observations show that
639 these laminae are composed mainly of fine grains with secondary sericite and Ti oxide (Figs. 6, 8,
640 and 12), and are aligned with the bedding and sometimes crosscut detrital grains and microquartz

641 cements that formed during diagenetic burial (Fig. 7). These observations suggest a strong
642 contribution of post-burial metasomatic fluid flow to the lamina during Mesoarchean to
643 Paleoproterozoic tectono-thermal events (Fig. 12c). Moreover, the multiple sulfur isotope records of
644 pyrite indicate the secondary addition of hydrothermal sulfur to the analyzed sandstones possibly via
645 fluid injections (Saitoh et al., 2020). Nonetheless, the major and trace element compositions of these
646 sandstones are not depleted in moderately immobile elements, and the MuQ-normalized REE
647 patterns show no positive Eu anomaly (Fig. 11). These geochemical data do not support an extensive
648 hydrothermal fluid circulation, but suggests that the fluids were rather endogenous and circulated
649 only within the sedimentary unit. The observed Moodies anastomosing and wavy laminae are thus
650 closely associated with low-grade fluid-mediated metamorphic events (Fig. 12). Similar dark
651 laminated textures with a mineral assemblage including Ti oxide were previously described in
652 younger 2.9 Ga sandstones of the Pongola Supergroup in Bower (2011), and were interpreted to be
653 formed as a result of the circulation of low-grade metamorphic fluids. The author precisely warned
654 the community about the risk to misinterpret lamination with such mineral assemblage as ancient
655 microbial mats.

656 Microscopic dark laminae from the same Moodies succession were previously referred as

657 “kerogenous” (Homann et al., 2015). However, carbonaceous matter was rarely identified in our
658 Raman analyses (Figs. 8 and 9). Our results demonstrate that not all wavy anastomosing laminae in
659 the Moodies rocks are kerogenous. Raman spectroscopy does not provide quantitative estimation of
660 the amount of carbonaceous matter and sufficient chemical information to identify disordered
661 carbonaceous matter as kerogen (Pasteris and Wopenka, 2003). However, both of the present and
662 previous studies reported the substantially low LOI (<2.55 wt.%) and TOC contents (<1000 ppm) in
663 the Moodies sandstones (Tables 2 and S2; Homann et al. 2018). It is worth noting that the lamina-
664 bearing subunits 1 and 4 do not show any TOC enrichment compared to the subunit 3 (devoid of
665 anastomosing and wavy lamination), suggesting that carbonaceous material is homogeneously scarce
666 in the studied Moodies sandstones (Table S2). Nonetheless, we emphasize that the present results do
667 not demonstrate that organic matter was ordinally scarce in the Moodies laminae in the subunits 1, 2,
668 and 4 during the sediment accumulation. Rather, it is likely that the observed fine-grained laminae
669 primarily contained some organic matter in the analyzed Moodies sediments (Stage 1) (Fig. 12a).
670 However, due to the substantial secondary overprints during the long post-depositional history (Fig.
671 12), it is difficult to determine whether the rarely detected carbonaceous materials in the laminae are
672 a remnant of original microbial mats or of sedimentary organic matter, which had been incorporated

673 into the fine-grained laminae during the sediment accumulation. In the analyzed Moodies
674 sandstones, intense silica precipitation that occurred mainly in the coarse-grained sediments after
675 burial (Stage 2) may have prevented, at least in part, organic remains from escaping from the
676 sediments during burial (Fig. 12b). Indeed, the entombment of microorganisms into silica
677 significantly limits their molecular degradation during experimental diagenesis (Alleon et al., 2016;
678 Igisu et al., 2018). Spatially resolved studies of organic microstructures in similarly metamorphosed
679 Paleoproterozoic cherts have consistently documented their chemical preservation with both
680 geochemical and petrographic evidence against organic migration from the microstructures (e.g., van
681 Zuilen et al., 2007; Westall et al., 2011; Alleon et al., 2018, 2021; Hickman-Lewis et al., 2020). On
682 the other hand, later metasomatism (Stage 3) and a recent oxidative weathering event (Stage 5) may
683 have participated to leach carbonaceous matter from the Moodies sediments (Petsch et al., 2000).

684 The occurrence of carbonaceous material in the Moodies sandstones has been reported in
685 several other localities in the central Saddleback Syncline (Gamper et al., 2011) and in the Stolzberg
686 and Dycedale synclines (Noffke et al. 2006; Nabhan et al., 2017; Homann et al., 2018; Köhler and
687 Heubeck, 2019), and has been associated with the morphology of anastomosing and wavy laminae
688 interpreted as a remnant of original microbial mats. However, based on the present petrological and

689 geochemical observations, the anastomosing and wavy laminations frequently observed in the
690 studied Moodies sandstones are not necessarily consistent with the existence of original microbial
691 mats. Our results also emphasize that the Moodies sediments are substantially different from cherts
692 from the older Onverwacht and Fig Tree groups that contain substantial amounts of organic matter
693 (e.g., Hofmann and Bolhar, 2007; Van Zuilen et al. 2007; Hofmann et al., 2013; Marin-Carbonne et
694 al. 2018; Alleon et al., 2021). In the future, research on new sedimentary successions and rock
695 samples would advance our understanding of traces of life preserved in the Moodies sediments,
696 especially in the framework of the upcoming BASE ICDP drilling project, although it will be
697 important to bear in mind the secondary overprint of metasomatism on the Moodies laminae.

698

699 **8. Conclusions**

700

701 The quartz-rich sandstones of the 3.22 Ga Moodies Group were collected from a ~350 m thick
702 succession in the Saddleback Syncline in the central Barberton Greenstone Belt, South Africa.

703 Petrological and multiple geochemical characteristics of the rocks were analyzed to reconstruct their
704 post-depositional history and to examine the origin of anastomosing and wavy laminae in the rocks,

705 which were previously suggested to be a remnant of microbial mats. The laminae are frequently
706 observed in coastal floodplain and inter- to supra-tidal settings, but not in a consistently high-energy
707 subtidal setting, in the analyzed succession. Our results show that, after an initial phase of
708 sedimentation (Stage 1), the studied sandstones have been affected by complex post-depositional
709 processes, including at least three main processes with four post-depositional stages: (i) intergranular
710 pressure solution leading to the microquartz cementation during the diagenesis burial (Stage 2); (ii)
711 formation of a sericite cement (Stage 3) and Ti oxide (Stage 4) during low-grade and fluid mediated
712 metamorphism; and (iii) rainwater infiltration that triggered more recent precipitation of Fe oxide
713 (Stage 5). This post-depositional history appears strongly dependent on the paleoenvironment and
714 nature of the sediments. In high energy settings, coarse-grained sediments characterized by an
715 intergrain porosity allowed the microquartz cementation, which prevented the coarse sediments from
716 later fluid injections. In contrast, under fluctuating energy settings, periods of more quiescent
717 hydrodynamic conditions allowed the repeated deposition of fine-grained laminae with a clay
718 matrix. This original matrix, which prevented the fine-grained laminae from the microquartz
719 cementation, was later replaced with sericite. Although we do not exclude that some of the observed
720 Moodies laminae were originally microbial mats, the mineral assemblage of the studied

721 anastomosing and wavy laminae (i.e., mainly sericite associated with Ti oxide and rare carbonaceous
722 material) shows a substantial metamorphic overprint, implying that similar laminae cannot be
723 unambiguously associated with the concept of a flourishing Paleoproterozoic microbial life at 3.22 Ga.

724

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737

738 **References**

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1058

1059 **Figure captions**

1060

1061 Fig. 1. Geological map (a) and stratigraphy (b, c) of the Moodies Group. (a) Simplified geological
1062 map of the Barberton Greenstone Belt (BGB) in northeast South Africa. The map is an enlarged
1063 square in the inset. A star shows the sampling locality in this study. (b) General stratigraphy of the
1064 Moodies Group in the Saddleback Syncline. (c) The analyzed sandstones in this study with
1065 sedimentary environments (according to Homman et al., 2015). Figures are modified from Homann
1066 et al. (2015).

1067

1068 Fig. 2. Outcrop photos of the analyzed Moodies sandstones. (a) A distant outcrop view of the
1069 sampling locality in the central Saddleback Syncline (the same hill of the stratigraphic Log 9 in
1070 Homann et al., 2015). The Moodies strata are dip subvertically and overturned. A person (circled) for
1071 scale. (b) Conglomerate (lower part) and megaripple (upper part) in the subunit 1. (c) Anastomosing
1072 and wavy laminae in the subunit 1. (d) Small channel, small megaripples, and flat laminae with

1073 ripples in the subunit 2. (e) Large-scale cross-bedding in the subunit 3. (f, g) gas-/fluid-escape
1074 structures in the subunit 4. A hammer (f) and pen (g) for scale are 28-cm- and 12-cm-long,
1075 respectively.

1076

1077 Fig. 3. Clastic grains of the analyzed sandstones. (a) Typical medium-grained quartz-rich sandstones
1078 (crossed nicols). (b) A quartz (Qz) grain with wavy extinction (crossed nicols). (c) A microcline
1079 (Mc) grain (crossed nicols). A chert (Ch) grain is also surrounded by quartz grains in the right side.
1080 (d) A rounded chert grain (crossed nicols). Arrows show intergranular pressure solution (IPS)
1081 texture. (e) Apatite inclusions (circled) in a quartz grain. Muscovite (Ms) is also shown. (f) BSE
1082 image of monazite-(Ce) (white grains). A partly albitized (Ab) K-feldspar (Kfs) is also shown. (g, h)
1083 EDS spectra of an apatite inclusion in quartz (g) and of monazite-(Ce) (h).

1084

1085 Fig. 4. Cements in the analyzed sandstones (crossed nicols except for d). (a) Coarse-grained
1086 sandstone with microquartz cements (arrows) in the subunit 3. (b) Fine-grained lamina enriched in
1087 sericite. (c) Boundary between lower coarse sediments and an upper fine-grained lamina enriched in
1088 sericite (intergranular brown crystal). (d) BSE image of elongated sericite (Ser) crystals between

1089 quartz (Qz) grains. (e, f) Sericite-bearing veins cutting irregular-shaped microquartz cements

1090 (arrows).

1091

1092 Fig. 5. Secondary minerals in the analyzed sandstones. (a) Reddish brown titanium (Ti) oxides

1093 (arrows) in grain boundaries (opened nicols). (b) BSE image of Ti oxides (arrow). (c-f) Ti oxide

1094 bearing vein (arrow) cutting a quartz grain (c, d) and microquartz-sericite cements (e, f). c and e:

1095 opened nicols, d and f: crossed nicols. (g) BSE image of iron oxides (arrows) along a micropore. (h)

1096 Pyrite particles (reflected light).

1097

1098 Fig. 6. Anastomosing and wavy laminae in the analyzed sandstones. (a, b) Outcrop (a) and slab (b)

1099 photos of the laminae. (c, d) A fine-grained microscopic lamina filled with sericite cements.

1100 Titanium oxide particles (black dots) are commonly observed within the lamina (arrows). c: opened

1101 nicols, d: crossed nicols. (e, f) Ti oxides (arrows) at the boundary between lower coarse-grained

1102 sediments and an upper fine-grained lamina. e: opened nicols, f: crossed nicols.

1103

1104 Fig. 7. Photomicrograph (opened nicols) and EPMA maps of a lamina in the subunit 4 of the

1105 analyzed succession. The lamina is composed mainly of fine-grained quartz and K-feldspar with
1106 Mg-bearing sericite and Fe- and Ti-oxides. Note that a larger chert grain at the lower left corner is
1107 cut by the lamina (arrows).

1108

1109 Fig. 8. Raman spectra of the analyzed laminae in SAD21 (a-c) and in SAD4 (d-f) in the subunit 1 of
1110 the analyzed succession. Flat/wavy laminae at the hand-specimen scale are composed mostly of Ti
1111 oxides (rutile and anatase). D1, G, and D2 bands of carbonaceous material (CM) was rarely detected
1112 (c) in the laminae for most of the samples investigated. Only some micrometric regions in one
1113 lamina in SAD39 are enriched sporadically in CM in the present data set (see Fig. 9).

1114

1115 Fig. 9. Raman maps on the CM-bearing lamina in SAD39. Note that this is the only lamina enriched
1116 sporadically in CM in the present dataset (also shown in Fig. 6c). (a) Microphotograph of the lamina
1117 (opened nicols). Lined and mapped regions are shown. On a line, a spot at which CM was detected is
1118 shown in red. Note that, even in this lamina, CM exists only sporadically. (b) Raman map 1. CM and
1119 K-feldspar are shown in red and yellow, respectively. (c-e) Raman map 2. Note an apparent
1120 correspondence between CM and Ti oxide. (f) Raman spectra showing the coexistence of CM and

1121 rutile on the map 1. Background fluorescence was subtracted.

1122

1123 Fig. 10. $\text{SiO}_2\text{-Al}_2\text{O}_3$ (a), $\text{K}_2\text{O-Al}_2\text{O}_3$ (b), $\text{Fe}_2\text{O}_3\text{-MgO}$ (c), and $\text{SiO}_2\text{-LOI}$ (d) cross plots of the
1124 analyzed Moodies succession. Previous sandstone data are from Hessler and Lowe (2006). Shale
1125 data are from McLennan and Taylor (1983), Hessler and Lowe (2006), and Toulkeridis et al. (2015).
1126 Igneous clast data are from Sanchez-Garrido et al. (2011) and Agangi et al. (2018). The present-day
1127 upper continental crust (UCC) composition is from Rudnick and Gao (2014).

1128

1129 Fig. 11. Chondrite- and MuQ-normalized REE patterns of the four subunits of the analyzed
1130 succession. Note that the REE patterns are generally consistent through the succession and no clear
1131 Eu anomaly is recognized on the MuQ-normalized patterns.

1132

1133 Fig. 12. Schematic model for the depositional and post-depositional history (in the five stages; a-e)
1134 of the analyzed Moodies sandstones. IPS: intergranular pressure solution.